Argonne
NATIONAL LABORATORY

ANL/NE-16/46

Van de Graaff Irradiation of Materials and Components during FY 2016

Nuclear Engineering Division

About Argonne National Laboratory
Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC under contract DE-AC02-06CH11357. The Laboratory's main facility is outside Chicago, at 9700 South Cass Avenue, Argonne, Illinois 60439. For information about Argonne and its pioneering science and technology programs, see www.anl.gov.

ANL/NE-16/46

Van de Graaff Irradiation of Materials and Components during FY 2016

by
Kevin Quigley, Sergey Chemerisov, Peter Tkac, and George F. Vandegrift
Nuclear Engineering Division, Argonne National Laboratory

October 2016

CONTENTS

FIGURES

TABLES

VAN DE GRAAFF IRRADIATION OF MATERIALS AND COMPONENTS DURING FY 2016

Kevin Quigley, Sergey Chemerisov, Peter Tkac, and George F. Vandegrift

October 2016

ABSTRACT

Through irradiations using our 3 MeV Van de Graaff accelerator, Argonne is testing the radiation stability of components of equipment that are being used to dispense molybdenum solutions for use as feeds to ^{99m}Tc generators and in the ^{99m}Tc generators themselves. Components have been irradiated by both a direct electron beam and photons generated from a tungsten convertor.

1 INTRODUCTION

NorthStar Medical Technologies, LLC (NorthStar), has developed the RadioGenix™ (formerly known as TechneGen) Generator System, which allows nuclear pharmacies to provide ^{99m}Tc for critical medical procedures using low-specific-activity ^{99}Mo as the source material. Molybdenum-99 is the parent isotope of ^{99m}Tc, an isotope used in approximately 85% of diagnostic imaging procedures. The RadioGenix™ System allows for the efficient separation and dose preparation of ^{99m}Tc from ^{99}Mo [MESSINA-2014]. The DPHARM is a ^{99m}Tc dispensing system, also known as the "^{99}Mo Aliquoting System." There is an interest in determining the lifetime of the components that are used in these systems. NorthStar performed calculations to predict the dose rates these components are expected to receive during operation. Argonne performed irradiations using a 3 MeV Van de Graaff electron accelerator (VDG). Components have been irradiated by both a direct electron beam and photons generated from a tungsten convertor. Components that will be in contact with solution in either the dispensing unit or the generator were returned to NorthStar, which performed leach tests to determine the radiation stability as required by the Federal Drug Administration (FDA).

2 CALCULATION OF DOSE RATES

NorthStar determined and provided Argonne the dose that each component is expected to receive during its lifetime. Calculations were based on the time that each component would be exposed to the fluids moving through the system. This value was multiplied by the number of runs each part was expected to see. Some parts are only used for ten elutions and replaced, while other parts in the system will be replaced annually. The time that the source material was in contact with each part and the dose expected from the source material determined the expected dose for each component.

3 ELECTRON BEAM PROFILE AND DOSE RATES

The electron irradiations were all performed with the 3 MeV electron beam; the irradiation was conducted at 25 in. from the window. Oxalic acid dosimetry was used to determine the dose (Figure 1) [QUIGLEY-2014]. The current profile moving perpendicularly from the center of the beam is reported in Figure 2. The corresponding variation of the dose within 1-in. distance from the center of the beam is less than 10%.

The X-ray irradiations were generated 12 in. from the beam window using a tungsten converter. This instrument has plates of 0.55-mm thick tungsten and 1.2-mm thick aluminum that convert the electron beam into X-rays (Figure 3). The dose at 12 in. using the converter vs. the current is shown in Figure 4. Doses were measured with a Radcal Corp. Model 9010 radiation monitor controller and a Radcal Model 9060A ion chamber.

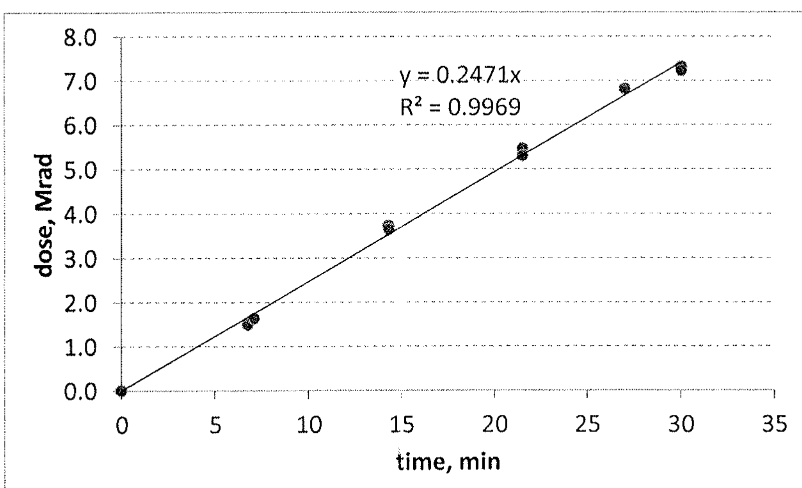

FIGURE 1 Linear relationship between the time and dose determined from the irradiations of 90-100 mL of 0.6 M oxalic acid at 20 µA electron beam current and 25 in. from the window.

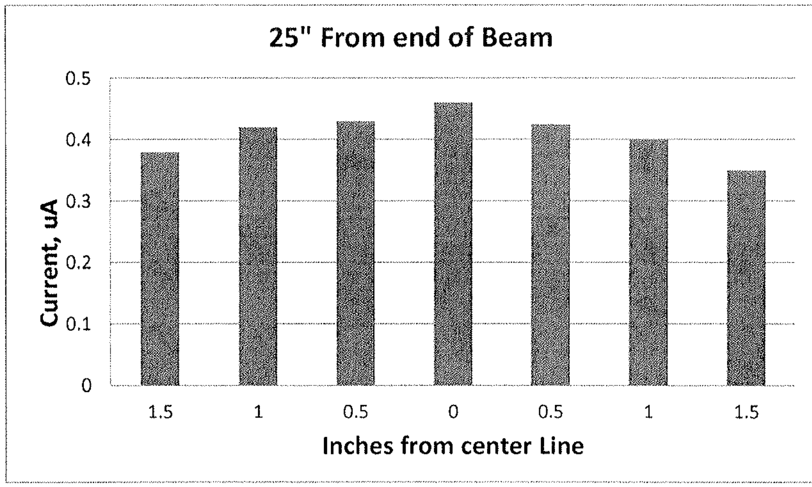

FIGURE 2 Profile of 10-μA beam current at 25 in. from window and various distances from center line using a 50-Ω target for direct electron beam irradiations.

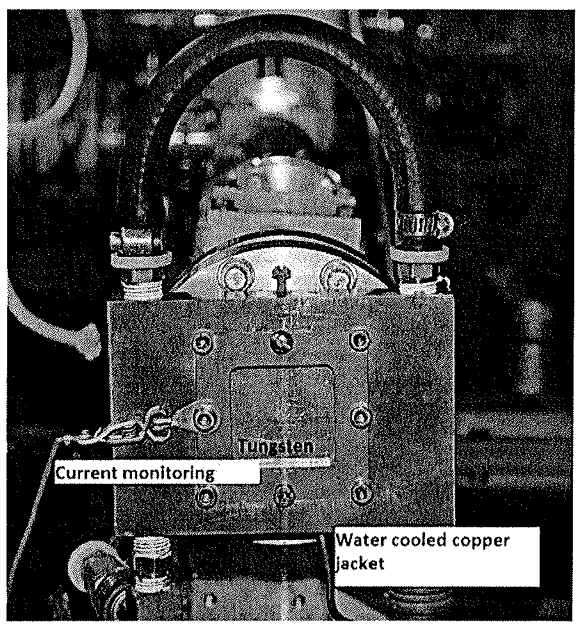

FIGURE 3 Tungsten converter with a water cooled-copper jacket

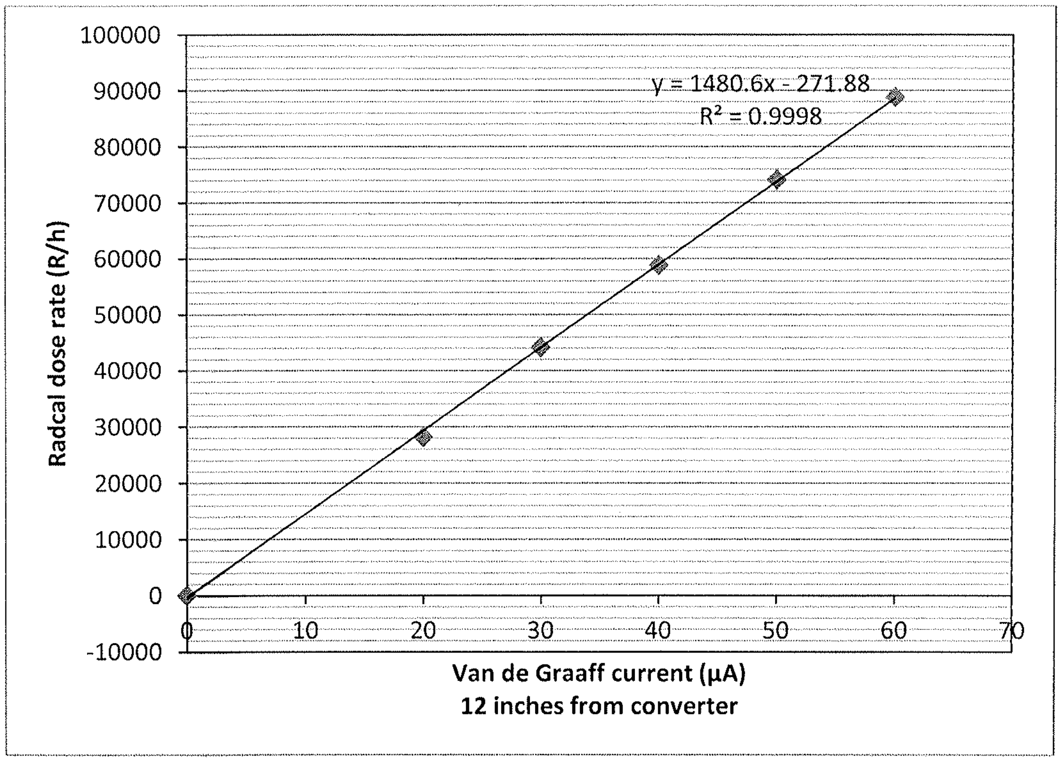

FIGURE 4 Linear relationship between the beam current and X-ray dose obtained 12 in. from tungsten converter and determined with a Radcal dosimeter

4 RESULTS

Syringes, O-rings, pressure sensors, and MVP4 controllers were irradiated during FY-16. The results are discussed below.

4.1 SYRINGES

Table 1 displays irradiations that were completed in FY-15, but the results of NorthStar's testing were not available for reporting earlier. Based on these results, further testing is underway. The conditions for these irradiations are shown in Table 2. NorthStar has changed the interface between the valves and syringes, and we will be irradiating two sets of syringes to see which interface has better radiation stability.

TABLE 1 X-ray irradiation of Hamilton syringes (part number 0203068)

Dose (kGy)	Dose Equivalent with Enriched-Mo Target	Number of Syringes	Results
9	3 month	2	2 passed
18	6 month	2	1 passed, 1 failed
27	9 months	2	2 failed
36	1 year	2	2 failed

TABLE 2 Repeat X-ray irradiation of Hamilton syringes (part number 0203068)

Dose (kGy)	Dose Equivalent with Enriched-Mo Target	Number of Syringes	Results
9	3 month	2	In progress
14	4.7 month	2	In progress
19	6 month	2	In progress
24	8 month	2	In progress

4.2 O-RINGS

NorthStar provided O-rings for electron irradiation at four dose rates—25 kGy, 50 kGy, 1 MGy, and 10 MGy. For the lower doses, the O-rings were irradiated by themselves; for the higher doses, they were irradiated with pressure sensors (discussed in the following section). Irradiations were completed for the three lower doses, and the irradiated samples were sent to NorthStar for testing. We have not yet learned the results of those tests. The 10-MGy irradiation is still underway. Figure 5 shows two bags of O-rings in a fixture with two pressure sensors before irradiation.

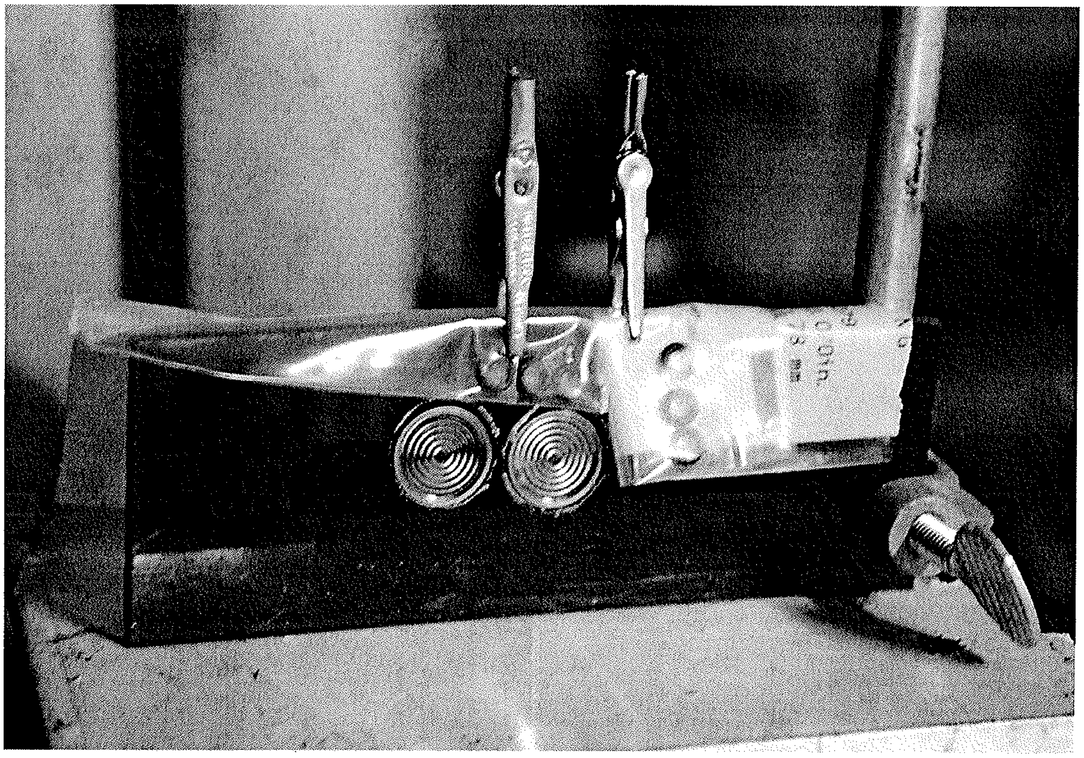

FIGURE 5 Original positioning of the pressure sensors and O-rings

4.3 PRESSURE SENSORS

Two pressure sensors (154B) supplied by NorthStar were to be irradiated to 1 and 10 MGy, or until failure. Figure 5 is a photograph of the sensors before irradiation. The pressure sensors were not monitored for electrical signal failure during this irradiation; they were to be shipped back to NorthStar and tested at the end of the irradiation. During this test, no cooling was used on the sensors, making it hard to determine if thermal stress caused or contributed to the physical failure seen in Figure 6. Physical failure of the two sensors occurred at an accumulated dose of 612 kGy, at a dose rate of 22.4 kGy/min. The diaphragms ruptured as shown in Figure 6. A thermocouple placed with the sensor read 86°C.

A second set of four irradiations was performed on the pressure sensors. In these irradiations, the sensors were connected to a computer and monitored to determine at what dose they no longer functioned. These components were cooled with compressed air. The sensors were irradiated to electrical failure. One sensor was irradiated to 800 kGy, and another to 1.5 MGy to determine if the diaphragm failure in the first test set would be repeated. Temperature during these tests averaged around 25.3°C. Neither of the sensors physically breached the diaphragm, as was seen in the first set. The four sensors tested thus far have failed to send electrical signals at 112 kGy, 107 kGy, 135 kGy, and 147 kGy. The first three irradiations had dose rates of 12.4 kGy/min, while the fourth irradiation was conducted at 22.4 kGy/min up to an accumulated dose of 1.5 MGy.

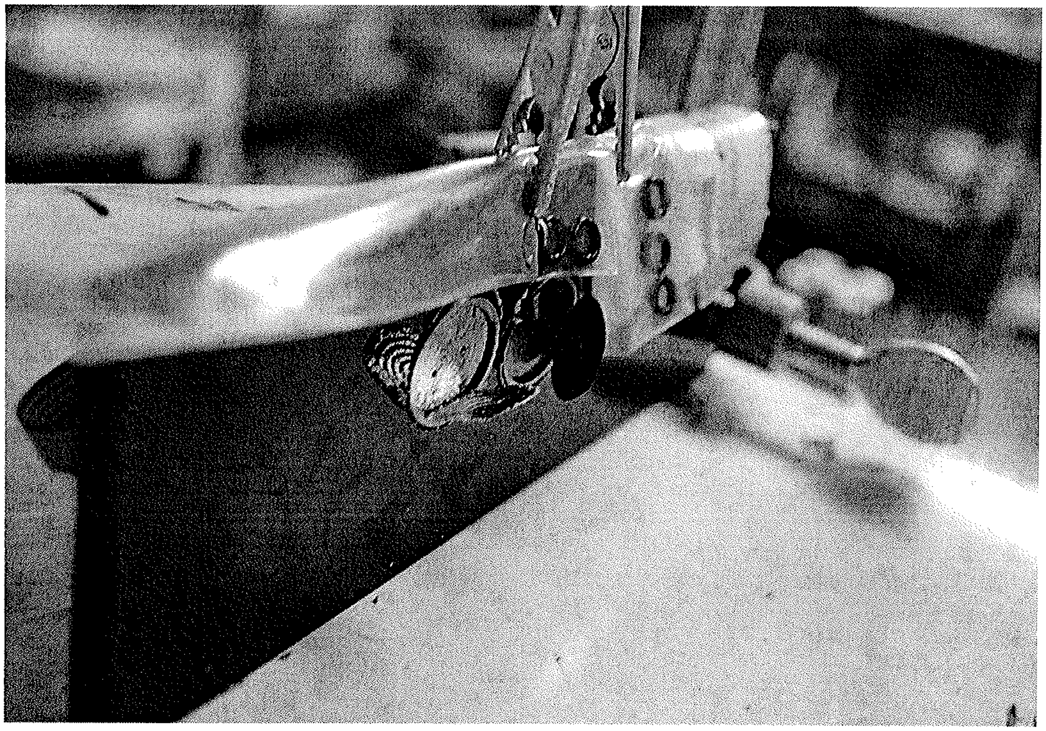

FIGURE 6 Rupture of the diaphragms at 612 kGy

These results indicate that the sensors will quit working at a dose far lower than the dose at which it was observed that the diaphragm breached. A fifth pressure sensor will be irradiated when we receive direction from NorthStar on how they want us to proceed.

4.4 MYP4 CONTROLLERS

The MVP4 controllers supplied by NorthStar were irradiated by X-rays using a tungsten photon converter. Table 3 shows the date and dose rate that each component was irradiated. The MVP4 controllers were hooked up to a 25-ft cable and run from a laptop during irradiations. They were monitored via a camera to see when failure occurred. Due to the unsatisfactory results shown in Table 3, NorthStar is having tungsten plates machined to shield the controllers. The controllers with the tungsten shields will be irradiated in the near future.

TABLE 3 Irradiations of MVP4 controllers using tungsten converter

Date	Controller Sample	Dose Rate (Gy/min)	Dose Applied (Gy)	Results
7/21/2016	MVP02GD1002 MVP4	15	360	Began grinding noise and failed
7/21/2016	MVP4 MVP02GD1016	7.5	360	Controller failed

5 CONCLUSIONS

Van de Graaff irradiations continue to be a useful tool for determining the radiation stability of materials and components. Irradiations by both electrons and X-rays are possible, and dose rates can be very high, especially for a focused electron beam; thus high doses can be achieved in a short time. The dose rate can be varied by changing the current.

6 REFERENCES

[MESSINA-2014] G.P. Messina, G.H. Isensee, J.T. Harvey, S.D. Moffatt, "New Generating System for Tc-99m Production," presented at the Mo-99 Topical Meeting, Washington, D.C., June 24-27, 2014, http://mo99.ne.anl.gov/2014/pdfs/presentations/S3P3%20Presentaton%20Harvey.pdf.

[QUIGLEY-2014] K. Quigley, V. Makarashvili, S. Chemerisov, G. Vandegrift, and P. Tkac, *Irradiation Stability of Syringe and Valve Controllers of RadioGenix System*, Argonne National Laboratory Report ANL/CSE-14/19 (July 2014).

Argonne
NATIONAL LABORATORY

Nuclear Engineering Division
Argonne National Laboratory
9700 South Cass Avenue, Bldg. 208
Argonne, IL 60439-4854

www.anl.gov

U.S. DEPARTMENT OF
ENERGY